



(19)

Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

EP 0 829 521 A1

(12)

## EUROPEAN PATENT APPLICATION

(43) Date of publication:  
18.03.1998 Bulletin 1998/12

(51) Int. Cl.<sup>6</sup>: C08L 101/02, C08L 83/04

(21) Application number: 97115853.0

(22) Date of filing: 11.09.1997

(84) Designated Contracting States:  
AT BE CH DE DK ES FI FR GB GR IE IT LI LU MC  
NL PT SE

(30) Priority: 11.09.1996 JP 240819/96

(71) Applicant: NEC CORPORATION  
Tokyo (JP)

(72) Inventors:  
• Serizawa, Shin  
Minato-ku, Tokyo (JP)  
• Iji, Masatoshi  
Minato-ku, Tokyo (JP)

(74) Representative:  
Glawe, Delfs, Moll & Partner  
Patentanwälte  
Postfach 26 01 62  
80058 München (DE)

### (54) Flame retardant resin composition

(57) A flame retardant resin composition comprises a silicone resin having specific structure and molecular weight and a non-silicone resin having an aromatic-ring. The silicone resin has first units of  $R_2SiO_{1.0}$  and second units of  $RSiO_{1.5}$ . The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. R is a radical selected from the group consisting of saturated or aromatic ring hydrocarbon groups.

EP 0 829 521 A1

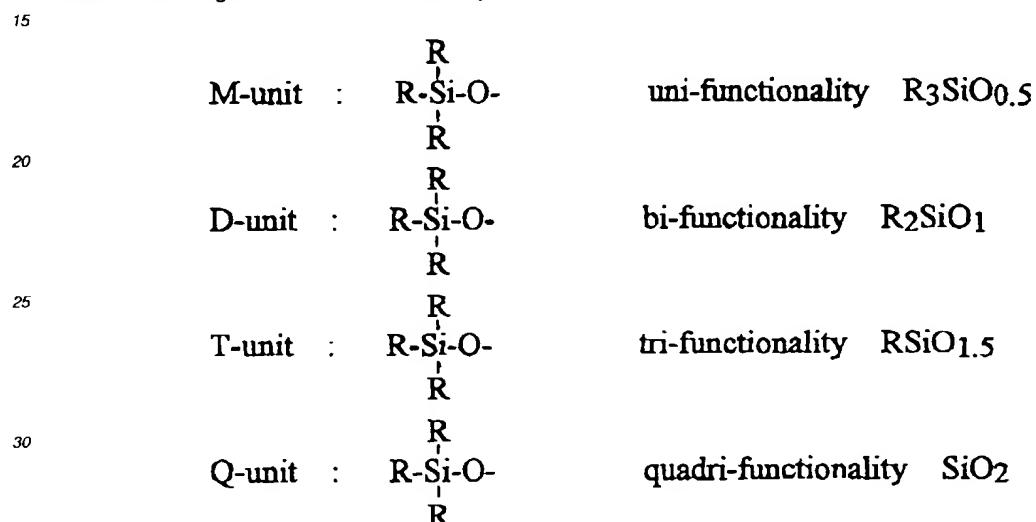
## Description

The present invention relates to a flame retardant resin composition, and more particularly to a silicone resin acting as a flame retarder which provides a flame retardancy to non-silicone resin component containing aromatic ring.

5 Flame retardant resin compositions have been widely used for electric and electronic components and devices, building materials, automobile parts, daily necessities and the like. In general, such base material resin components are added with an organic halogen compound or added with both an organic halogen compound and antimony trioxide so that the base material resin components are provided with the flame retardancy. Such the flame retardant resin material has a disadvantage that burning this flame retardant resin material causes generation of harmful halogen based gases.

10 It has been known by one skilled in the art that in order to avoid the above disadvantage it is effective to add the base material resin component with silicone thereby to provide the flame retardancy to the base material resin component without generation harmful halogen based gases when burning.

Silicone (organopolysiloxane) comprises at least any of the following four units (M-unit, D-unit, T-unit and Q-unit). Silicone resin in general includes at least any of T-unit and Q-unit.



In the Japanese laid-open patent publication No. 1-318069, it is disclosed that a thermoplastic material is provided with a flame retardancy by powdered polymer mixture which comprises a thermoplastic polymer and a silicone resin ( $\text{R}_x\text{Si}(\text{OR}')_y\text{O}_{(4-x-y)/2}$ ) consisting of all of the above four units and also having either alkoxide group or hydroxyl group.

40 In the Japanese laid-open patent publication No. 2-150436, it is disclosed that a thermoplastic material is provided with a flame retardancy by a mixture of the normal silicone powder such as dimethyl-silicone, metal hydroxide and zinc compound.

45 In the Japanese patent publication No. 62-60421, it is disclosed that a thermoplastic non-silicone polymer is provided with a flame retardancy by polysiloxane containing 80 % by weight or more of T-unit represented by  $\text{RSiO}_{1.5}$ . Particularly, it is preferable for providing the flame retardant to the polymer component that the molecular weight of the polysiloxane resin is not less than 2000 and not more than 6000 and radical R in the formula comprises not more than 80mol% of phenyl group and the remaining percents of methyl group.

50 In the Japanese patent publication No. 3-48947, it is disclosed that a thermoplastic material is provided with a flame retardancy by a silicone resin, particularly a MQ silicone which comprises M-unit of  $\text{R}_3\text{SiO}_{0.5}$  and Q-unit of  $\text{SiO}_2$ , silicone and Group IIA metal salts.

55 In the Japanese laid-open patent publication No. 1-318069 or No. 2-150436, the use of normal silicone resin alone results in an insufficient flame retardancy, for which reason flame retarder such as halogen based compounds, phosphorus based compounds, metal hydroxide may be used and added.

The specific silicone resins disclosed in the Japanese patent publications Nos. 62-60421 and 3-48947 have to be added at a large amount for obtaining a sufficient flame retardancy

60 In the Japanese patent publication No. 62-60421, 10 parts by weight to 300 parts by weight of silicone resin, preferably 20 parts by weight to 100 parts by weight, are added to 100 parts by weight of non-silicone resin for flame retardancy. It is however disadvantageous that addition of such large amount of silicone resin results in remarkable

deterioration of formability and mechanical strength of the resin component.

In the above circumstances, it had been required to develop a flame retardant resin composition having a high flame retardancy with addition of a small amount of silicone resin, and also having high formability and mechanical strength.

5 Accordingly, it is an object of the present invention to provide a novel flame retardant resin composition free from the above problems and disadvantages.

It is a further object of the present invention to provide a novel flame retardant resin composition having a high flame retardancy with addition of a small amount of silicone resin.

10 It is a still further object of the present invention to provide a novel flame retardant resin composition having a high formability.

It is yet a further object of the present invention to provide a novel flame retardant resin composition having and a high mechanical strength.

It is yet a further object of the present invention to provide a novel flame retardant resin composition which generates almost no harmful halogen based gases when burning this flame retardant resin composition.

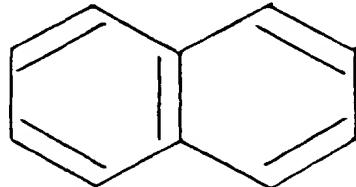
15 The above and other objects, features and advantages of the present invention will be apparent from the following descriptions.

The present invention provides a novel flame retardant resin composition which comprises a silicone resin having specific structure and molecular weight and a non-silicone resin having an aromatic-ring. The silicone resin has first units of  $R_2SiO_{1.0}$  and second units of  $RSiO_{1.5}$ . The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. R is a radical selected from 20 the group consisting of saturated or aromatic ring hydrocarbon groups.

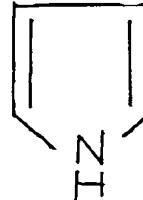
25 The present invention provides a novel flame retardant resin composition which comprises a silicone resin having specific structure and molecular weight and a non-silicone resin having an aromatic-ring. The silicone resin has first units of  $R_2SiO_{1.0}$  and second units of  $RSiO_{1.5}$ . The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. R is a radical selected from the group consisting of saturated or aromatic ring hydrocarbon groups.

The above aromatic ring may be rings which fall into aromatic rings such as benzene ring, condensed benzene ring, non-benzene system aromatic ring, complex aromatic ring. The following naphthalene ring is one example of the condensed benzene ring and the following payroll ring is one example of the complex aromatic ring.

30



35



payroll ring

40

The above non-silicone resin may be thermoplastic resins having aromatic ring such as aromatic system polycarbonate resin, alloys of aromatic system polycarbonate resin, copolymers of acrylonitrile-butadiene-styrene (ABS) and 45 polystyrene resin and also may be thermoplastic resin having aromatic ring such as epoxy resin having aromatic ring and phenol resin having aromatic ring. Further, the above non-silicone resin may be a mixture of a plurality of the above thermoplastic resins. Aromatic polycarbonate resin is preferable for providing flame retardancy.

As described above, the silicone resin has first units of  $R_2SiO_{1.0}$  referred to as D-unit and second units of  $RSiO_{1.5}$  referred to as T-unit. R is a radical selected from the group consisting of saturated or aromatic ring hydrocarbon groups. 50 The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. If the molecular weight of the silicone resin is less than 10000, then it is difficult to mix the silicon resin into the base material, resulting in insufficient flame retardancy effect. If, however, the molecular weight of the silicone resin is extremely high, then an excessively high melting viscosity is obtained thereby lowering the mixability and formability of the silicone resin with the non-silicone. For example, the preferable molecular weight may 55 be in the range of 10000 to 300000.

A preferable molar ratio of the first unit  $R_2SiO_{1.0}$  to the second units of  $RSiO_{1.5}$  may be in the range of 0.5 to 7. It is preferable that the first unit  $R_2SiO_{1.0}$  is included in the silicone resin in the range of 15% by weight to 75 % by weight. If the first unit  $R_2SiO_{1.0}$  is included in the silicone resin at less than 15% by weight, then it is difficult to mix the silicon

resin into the non-silicon resin as the base material and also then the flame retardancy is deteriorated. If, however, the first unit  $R_2SiO_{1.0}$  is included in the silicone resin at less than 15% by weight, then the flame retardancy is deteriorated.

5 Radical R in the first and second units  $R_2SiO_{1.0}$  and  $RSiO_{1.5}$  may preferably be methyl groups and phenyl groups. More preferably, a molar ratio of phenyl groups to the radical R in the first and second units  $R_2SiO_{1.0}$  and  $RSiO_{1.5}$  may be in the range of 20% to 80%. If the molar ratio of phenyl groups to the radical R in the first and second units  $R_2SiO_{1.0}$  and  $RSiO_{1.5}$  is less than 20%, then it is difficult to mix the silicon resin into the non-silicon resin as the base material and also then the flame retardancy is deteriorated. If, however, the molar ratio of phenyl groups to the radical R in the first and second units  $R_2SiO_{1.0}$  and  $RSiO_{1.5}$  is more than 80%, then the flame retardancy is deteriorated.

10 It is preferable that the bridging terminal group of the silicone resin is terminated or ended with  $R_2SiO_{0.5}$  and R' is methyl group or phenyl group.

It is possible that a single kind of the silicone resin is mixed to a singe kind of the non-silicone resin.

It is also possible that a plurality of kinds of the silicone resin is mixed to a singe kind of the non-silicone resin.

It is also possible that a single kind of the silicone resin is mixed to a plurality of kinds of the non-silicone resin.

15 It is also possible that a plurality of kinds of the silicone resin is mixed to a plurality of kinds of the non-silicone resin.

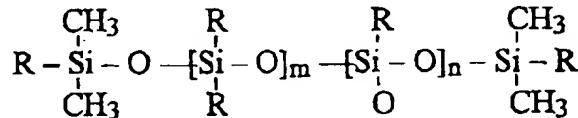
It is possible to add a reinforcing agent into the flame retardant resin composition. The reinforcing agent may be antioxidant, neutralizer, ultraviolet ray absorbent, anti-static agent, pigment, dispersing agent, lubricant, thickener, filler and the others mixable in the resin component.

20 The following descriptions will focus on a method of preparing the above flame retardant resin composition in accordance with the present invention.

25 As the non-silicone resin component having the aromatic ring, used are aromatic system polycarbonate resin, alloys of aromatic system polycarbonate resin, copolymers of acrylonitrile-butadiene-styrene (ABS) and polystyrene resin and also may be thermoplastic resin having aromatic ring such as epoxy resin having aromatic ring and phenol resin having aromatic ring. Further, the above non-silicone resin may be a mixture of a plurality of the above thermoplastic resins.

30 The silicone resin component may be prepared by the general method. Namely, in accordance with the molecular weight of the silicon resin component and a ratio of the first unit  $R_2SiO_{1.0}$  to the second unit  $RSiO_{1.5}$ , diorganochlorosilane and monoorganochlorosilane are hydrolyzed and partially condensed to form a silicone resin. Further, the silicone resin is then reacted with triorganoydroxysilane to terminate polymerization whereby the silicone resin is terminated with the bridging terminal group of  $R_3SiO_{0.5}$ .

35 The property of the silicone resin component depends upon the molecular weight, a ratio of the first unit to the second unit and a ratio of phenyl group to methyl group in the radical R. The general formula of the silicon resin is as follows.



40

The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. The molecular weight of the silicone resin is controllable by controlling a time of reaction for preparing the silicone resin.

45 The ratio of the first unit  $R_2SiO_{1.0}$  to the second unit  $RSiO_{1.5}$  in the silicone resin is adjustable by amounts of diorganochlorosilane and monoorganochlorosilane. Chlorine in the raw material for the silicone resin is made into hydrochloric acid during the hydrolyzing reaction whereby no chlorine is included in the silicone resin. The preferable molar ratio of the first unit  $R_2SiO_{1.0}$  to the second units of  $RSiO_{1.5}$  is in the range of 0.5 to 7. Namely, it is preferable that the first unit  $R_2SiO_{1.0}$  is included in the silicone resin in the range of 15% by weight to 75 % by weight. A ratio of the first unit to the second unit and a ratio of phenyl group to methyl group in the radical R is adjustable by amounts in use of both methyl-silane system raw material such as dimethylchlorosilane and monomethylchlorosilane and phenyl silane system raw material such as diphenylchlorosilane and monophenylchlorosilane.

50 The non-silicone resin component and the silicon resin component are determined and mixed with each other to be formed by the same apparatus as when used to form rubber plastic material. Namely, mixer and stiller such as ribbon blender are used to mix and disperse the non-silicone resin component and the silicon resin component separately before a milling and mixing device such as Bumbury's roll and extruder to mix the non-silicone resin component and the silicon resin component to form the intended material. If the silicone resin is not solid in the ordinary temperature, then the silicone resin is mixable with the non-silicone resin by the milling mixer.

After mixing the silicon resin to the non-silicone resin, the mixture is defined by injection molding, extrusion molding, condensation molding and vacuum molding thereby to obtain the flame retardant resin material.

After molding, in accordance with JIS K7201(burning examination of polymer material in oxygen index, the evaluation on flame retardancy is made. The burning examination of polymer material in oxygen index means a method for confirming whether nor not a predetermined size sample peace is continuously burned for a time not less than 3 minutes and also determining a minimum flow rate of oxygen and a corresponding nitrogen flow rate both of which are necessary for continuing the flame length of not less than 50 mm. The confirmed minimum oxygen concentration is oxygen index. As the oxygen index is high, then the flame retardancy is high.

As described above, the novel flame retardant resin composition comprises a silicone resin having specific structure and molecular weight and a non-silicone resin having an aromatic-ring. The silicone resin has first units of  $R_2SiO_{1.0}$  and second units of  $RSiO_{1.5}$ . The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. R is a radical selected from the group consisting of saturated or aromatic ring hydrocarbon groups. The novel flame retardant resin composition in accordance with the present invention has a high flame retardancy with addition of a small amount of silicone resin without, however, generation of harmful halogen based gases when burning this flame retardant resin composition.

Example 1:

Fifteen kinds of silicone resins "a", "b", "c", "d", "e", "f", "g", "h", "i", "j", "k", "l", "m", "n" and "o" were synthesized by varying the molecular weight of the silicone resin and the ratio of the first unit  $R_2SiO_{1.0}$  to the second unit of  $RSiO_{1.5}$  as well as the ratio of phenyl group to methyl group in the hydrocarbon group R. The radical R including the bridging terminal group comprise phenyl groups and methyl group. The following table 1 shows the molecular weight of the silicone resin and the ratio of the first unit  $R_2SiO_{1.0}$  to the second unit of  $RSiO_{1.5}$  as well as the ratio of phenyl group to methyl group in the hydrocarbon group R for every silicone resins "a", "b", "c", "d", "e", "f", "g", "h", "i", "j", "k", "l", "m", "n" and "o".

TABLE 1

Resin	Molecular Weight	First-To-Second Unit Ratio	Phenyl-To-Methyl Ratio
"a"	5,000	0:1	70:30
"b"	6,000	1:1	25:75
"c"	8,000	1:0.4	70:30
"d"	12,000	0:1	00:100
"e"	40,000	1:0	40:60
"f"	12,000	1:0.41	15:85
"g"	25,000	1:7.5	85:15
"h"	50,000	1:7.5	85:15
"i"	12,000	1:0.5	15:85
"j"	50,000	1:6.5	85:15
"k"	12,000	1:0.5	50:50
"l"	40,000	1:1	40:60
"m"	50,000	1:3	75:25
"n"	120,000	1:6.5	25:75
"o"	300,000	1:3	60:40

The novel flame retardant resin composition comprises a silicone resin having specific structure and molecular weight and a non-silicone resin having an aromatic-ring. The silicone resin has first units of  $R_2SiO_{1.0}$  and second units of  $RSiO_{1.5}$ . The molecular weight of the silicone resin is not less than 10000 provided that the silicone resin is mixable with the non-silicone resin having the aromatic-ring. R is a radical selected from the group consisting of saturated or

aromatic ring hydrocarbon groups. The silicone resins "f", "g", "h", "i", "j", "k", "l", "m" and "n" are the resin to be used in the novel flame retardant resin composition in the preferred examples according to the present invention. By contrast, the silicone resins "a", "b", "c", "d", "e" and "o" are the resin to be used in the flame retardant resin composition in the comparative examples. The silicon "o" is too high in molecular weight and melting viscosity to be mixed with the non-silicone resin, for example, polycarbonate resin.

As the non-silicone resin, used was bis-phenol A-polycarbonate resin (Sumitomo Dou Co - Caliber 301-10) or alloys of bis-phenol A-polycarbonate resin (Sumitomo Dou Co - Caliber 301-10) and ABS (Sumitomo Dou Co - H-270, hereinafter referred to as PC/ABS), or polystyrene resin (Shinnetsu Chemical H-65). The silicone resin and the non-silicone resin are milled and mixed by extruder stone gist mill at a temperature of 280°C when polycarbonate resin is used or at a temperature of 260°C when PC/ABS is used or at a temperature of 220°C when polystyrene resin is used.

The milled and mixed resin composition was dried at a temperature of 270°C when the polycarbonate is used or at a temperature of 240°C when PC/ABS system is used, or at a temperature of 200°C when the polystyrene system is used.

The formed plate is processed to a sample piece of a length 150 mm, a width of 6.5±0.5 mm so that in accordance with JIS K7201(burning examination of polymer material in oxygen index, the evaluation on flame retardancy is made to find oxygen index.

The following Table 2 shows an amount of the silicon resin with reference to 100 parts by weight of the non-silicone resin and an oxygen index for every resin compositions in preferred examples according to the present invention.

The following Table 3 shows an amount of the silicon resin with reference to 100 parts by weight of the non-silicone resin and an oxygen index for various resin compositions in comparative example when no silicone resin is added or when the molecular weight is less than 10000 or when the silicone resin does not have both the first and second units.

TABLE 2

	Example 1	Example 2	Example 3
Polycarbonate (non-silicone)	100	100	100
Silicone resin "f"	4	-	-
Silicone resin "g"	-	4	-
Silicone resin "h"	-	-	4
Evaluation (oxygen index)	31	30	31
(Unit :parts by weight)			

35

40

45

50

55

TABLE 3

	Com.Ex. 1	Com.Ex. 2	Com.Ex. 3	
5	Polycarbonate (non-silicone)	100	100	100
	Silicone resin "a"	-	4	-
10	Silicone resin "b"	-	-	4
	Silicone resin "c"	-	-	-
15	Silicone resin "d"	-	-	-
	Silicone resin "e"	-	-	-
	Evaluation (oxygen index)	26	27	28
	(Unit :parts by weight)			
20		Com.Ex. 4	Com.Ex. 5	Com.Ex. 6
	Polycarbonate (non-silicone)	100	100	100
	Silicone resin "a"	-	-	-
25	Silicone resin "b"	9	-	-
	Silicone resin "c"	-	4	-
	Silicone resin "d"	-	-	4
	Silicone resin "e"	-	-	-
30	Evaluation (oxygen index)	29	27	27
	(Unit :parts by weight)			
35		Com.Ex. 7		
	Polycarbonate (non-silicone)	100		
	Silicone resin "a"	-		
40	Silicone resin "b"	-		
	Silicone resin "c"	-		
	Silicone resin "d"	-		
	Silicone resin "e"	4		
	Evaluation (oxygen index)	26		
45	(Unit :parts by weight)			

From Tables 2 and 3, it is understood that the polycarbonate resin compositions containing the silicone resins "f", "g", "h" in Examples 1, 2 and 3 are high in oxygen index than the polycarbonate resin compositions containing the silicone resins "a", "b", "c", "d", "e" in Comparative Examples 2-7 and also than the polycarbonate resin free of silicone resin in Comparative Example 1. The flame retardant resin compositions of Examples 1, 2 and 3 are higher in flame retardancy than Comparative Examples 1-7.

50 The following Table 4 shows dependency of the oxygen index upon the ratio of the first unit to the second unit and the ratio of methyl group and phenyl group.

TABLE 4

		Example 4	Example 5	Example 6
5	Polycarbonate (non-silicone)	100	100	100
	Silicone resin "i"	4	-	-
	Silicone resin "j"	-	4	-
	Silicone resin "k"	-	-	4
	Silicone resin "l"	-	-	-
	Silicone resin "m"	-	-	-
	Silicone resin "n"	-	-	-
10	Evaluation (oxygen index)	32	33	36
	(Unit :parts by weight)			
15		Example 7	Example 8	Example 9
	Polycarbonate (non-silicone)	100	100	100
	Silicone resin "i"	-	-	-
	Silicone resin "j"	-	-	-
	Silicone resin "k"	-	-	-
	Silicone resin "l"	4	9	-
	Silicone resin "m"	-	-	4
	Silicone resin "n"	-	-	-
	Evaluation (oxygen index)	35	37	35
	(Unit :parts by weight)			
20		Example 10		
	Polycarbonate (non-silicone)	100		
	Silicone resin "i"	-		
	Silicone resin "j"	-		
	Silicone resin "k"	-		
	Silicone resin "l"	-		
	Silicone resin "m"	-		
	Silicone resin "n"	4		
	Evaluation (oxygen index)	34		
	(Unit :parts by weight)			
25		Example 11		
	Polycarbonate (non-silicone)	100		
	Silicone resin "i"	-		
	Silicone resin "j"	-		
	Silicone resin "k"	-		
	Silicone resin "l"	-		
	Silicone resin "m"	-		
30		Example 12		
	Polycarbonate (non-silicone)	100		
	Silicone resin "i"	-		
	Silicone resin "j"	-		
35		Example 13		
	Polycarbonate (non-silicone)	100		
	Silicone resin "i"	-		
	Silicone resin "j"	-		
	Silicone resin "k"	-		
40		Example 14		
	Polycarbonate (non-silicone)	100		
	Silicone resin "i"	-		
	Silicone resin "j"	-		
	Silicone resin "k"	-		
45		Example 15		
	Polycarbonate (non-silicone)	100		
	Silicone resin "i"	-		
	Silicone resin "j"	-		
	Silicone resin "k"	-		

From Examples 4 and 5, it is understood that the polycarbonate resin compositions containing the silicone resins

50 "i", "j" in Examples 4 and 5 are high in oxygen index than the polycarbonate resin compositions containing the silicone resins in Examples 1-3 and in Comparative Examples 1-7. The flame retardant resin compositions of Examples 4 and 5 are higher in flame retardancy than Examples 1-3 and Comparative Examples 1-7.

From Examples 6-10, it is understood that the polycarbonate resin compositions containing the silicone resins "k", "l", "m", "n" in Examples 6-10 are high in oxygen index than the polycarbonate resin compositions containing the silicone resins in Examples 1-5 and in Comparative Examples 1-7. The flame retardant resin compositions of Examples 6-10 are higher in flame retardancy than Examples 1-5 and Comparative Examples 1-7.

The following Table 5 shows dependency of the oxygen index when PC/ABS is used.

TABLE 5

	Example 11	Example 12	Example 13	
5	PC/ABS (non-silicone)	100	100	100
10	Silicone resin "b"	-	-	-
15	Silicone resin "d"	-	-	-
20	Silicone resin "T"	4	-	-
25	Silicone resin "T"	-	9	-
30	Silicone resin "T"	-	-	9
35	Evaluation (oxygen index)	23	25	26
40	(Unit :parts by weight)			
45		Com.Ex. 8	Com.Ex. 9	Com.Ex. 10
50	PC/ABS (non-silicone)	100	100	100
55	Silicone resin "b"	-	9	-
60	Silicone resin "d"	-	-	4
65	Silicone resin "T"	-	-	-
70	Silicone resin "T"	-	-	-
75	Silicone resin "T"	-	-	-
80	Evaluation (oxygen index)	19	20	19
85	(Unit :parts by weight)			

From Examples 11-13, it is understood that the polycarbonate resin compositions containing the silicone resins in Examples 11-13 are high in oxygen index than the polycarbonate resin compositions containing the silicone resins in Comparative Examples 8-10. The flame retardant resin compositions of Examples 11-13 are higher in flame retardancy than Comparative Examples 8-10.

The following Table 6 shows dependency of the oxygen index when polycarbonate is used.

TABLE 6

		Example 14	Example 15	Example 16
5	Polycarbonate (non-silicone)	100	100	100
	Silicone resin "b"	-	-	-
	Silicone resin "d"	-	-	-
	Silicone resin "f"	4	-	-
	Silicone resin "i"	-	9	-
	Silicone resin "l"	-	-	9
10	Evaluation (oxygen index)	22	24	25
	(Unit :parts by weight)			
15		Com.Ex. 11	Com.Ex. 12	Com.Ex. 13
	Polycarbonate (non-silicone)	100	100	100
	Silicone resin "b"	-	9	-
	Silicone resin "d"	-	-	4
	Silicone resin "f"	-	-	-
	Silicone resin "i"	-	-	-
	Silicone resin "l"	-	-	-
20	Evaluation (oxygen index)	18	19	18
	(Unit :parts by weight)			
25		Com.Ex. 11	Com.Ex. 12	Com.Ex. 13
	Polycarbonate (non-silicone)	100	100	100
30	Silicone resin "b"	-	9	-
	Silicone resin "d"	-	-	4
35	Silicone resin "f"	-	-	-
	Silicone resin "i"	-	-	-
40	Silicone resin "l"	-	-	-
	Evaluation (oxygen index)	18	19	18
45	(Unit :parts by weight)			

From Examples 14-16, it is understood that the polycarbonate resin compositions containing the silicone resins in Examples 14-16 are high in oxygen index than the polycarbonate resin compositions containing the silicone resins in Comparative Examples 11-13. The flame retardant resin compositions of Examples 14-16 are higher in flame retardancy than Comparative Examples 11-13.

Whereas modifications of the present invention will be apparent to a person having ordinary skill in the art, to which the invention pertains, it is to be understood that embodiments as shown and described by way of illustrations are by no means intended to be considered in a limiting sense. Accordingly, it is to be intended to cover by claims all modifications which fall within the spirit and scope of the present invention.

#### 40 Claims

1. A flame retardant resin composition which comprises a silicone resin and a non-silicone resin having an aromatic ring, characterized in that said silicone resin has first units of  $R_2SiO_{1.0}$  and second units of  $RSiO_{1.5}$  and a molecular weight of said silicone resin is not less than 10000 provided that said silicone resin is mixable with the non-silicone resin having the aromatic-ring and also R is a hydrocarbon group.
2. The flame retardant resin composition as claimed in claim 1, characterized in that said non-silicone resin comprises at least one selected from the group consisting of aromatic system polycarbonate resins, alloys of aromatic system polycarbonate resin, copolymers of acrylonitrile-butadiene-styrene, polystyrene resins, epoxy resins having aromatic ring and phenol resins having aromatic ring.
3. The flame retardant resin composition as claimed in claim 1, characterized in that said molecular weight is in the range of 10000 to 300000.
4. The flame retardant resin composition as claimed in claim 1, characterized in that a molar ratio of said first unit  $R_2SiO_{1.0}$  to said second units of  $RSiO_{1.5}$  is in the range of 0.5 to 7.

5. The flame retardant resin composition as claimed in claim 1, characterized in that R comprises a methyl group and a phenyl group.
6. The flame retardant resin composition as claimed in claim 5, characterized in that a molar ratio of said phenyl group to R is in the range of 20% to 80%.
7. The flame retardant resin composition as claimed in claim 1, characterized in that a bridging terminal group of said silicone resin is terminated with  $R'_2SiO_{0.5}$  and R' is one selected from the groups consisting of methyl groups and phenyl groups.
- 10 8. The flame retardant resin composition as claimed in claim 1, characterized in that a single kind of said silicone resin is mixed to a single kind of said non-silicone resin.
- 15 9. The flame retardant resin composition as claimed in claim 1, characterized in that a plurality of kinds of said silicone resin is mixed to a single kind of said non-silicone resin.
10. The flame retardant resin composition as claimed in claim 1, characterized in that a single kind of said silicone resin is mixed to a plurality of kinds of said non-silicone resin.
- 20 11. The flame retardant resin composition as claimed in claim 1, characterized in that a plurality of kinds of said silicone resin is mixed to a plurality of kinds of said non-silicone resin.
12. The flame retardant resin composition as claimed in claim 1, characterized in that a reinforcing agent is added into said flame retardant resin composition.
- 25 13. The flame retardant resin composition as claimed in claim 11, characterized in that said reinforcing agent is at least one selected from the group consisting of antioxidants, neutralizers, ultraviolet ray absorbents, anti-static agents, pigments, dispersing agents, lubricants, thickeners and fillers.

30

35

40

45

50

55



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 97 11 5853

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	EP 0 415 070 A (GENERAL ELECTRIC) * page 3, line 9 - line 40 * * page 3, line 56 - line 58 * * example *	1-13	C08L101/02 C08L83/04
X	US 5 391 594 A (D. J. ROMENESKO ET AL.) * column 4, line 53 - line 65 * * column 5, line 13 - line 16 * * column 5, line 40 - line 45 * * examples 1,2 * * tables 1,2 *	1-13	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C08L
<p>The present search report has been drawn up for all claims</p>			
Place of search	Date of completion of the search	Examiner	
BERLIN	17 December 1997	Hoepfner, W	
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons S : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			